Lawrence Livermore National Laboratory

Spiked Alloy Production

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Advances in Materials Science

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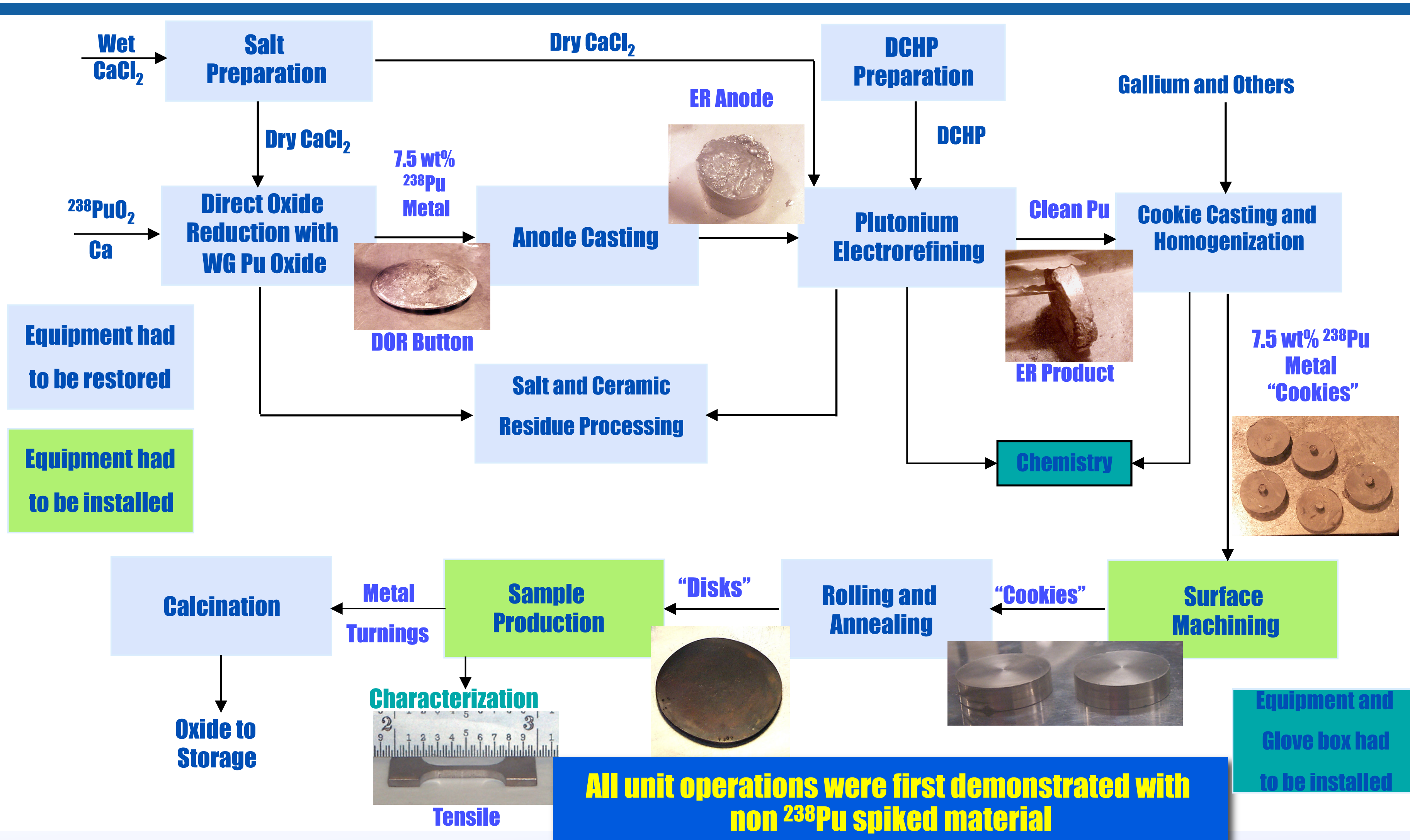
Collaborators

- J. A. McNeese, K. E. Dodson, W. L. Williams, O. H. Krikorian, M. S. Blau, J. E. Schmitz, F. G. Bajao, D. A. Mew
  - Plutonium Processing
- T. E. Matz
  - Machining
- R. A. Torres, K. J. Moody, J. M. Kenneally
  - Chemical and Isotopic Analysis

Background and Objective

- Plutonium metal in our nuclear stockpile is aging with time because the plutonium undergoes alpha decay, which leads to structural damage in the material.
  - The primary aging effects are believed to be helium bubble formation and void swelling.
- In order to study the accelerated aging of plutonium, an isotope of plutonium with a much higher activity, ^{238}Pu (also an alpha emitter), is used to spike plutonium alloys.
  - The aging in a 7.5% ^{238}Pu-spiked plutonium alloy is about 16 times faster than in weapons grade plutonium.
- By monitoring the aging of ^{238}Pu-spiked plutonium alloys for just a few years, we will be able to predict the aging behavior of plutonium in nuclear weapons over periods of 50 years or more.

^{238}Pu spiked alloy test specimens are produced to accelerate the effects of aging on plutonium



The spiked alloy is produced by calcium reduction of plutonium oxide

- PuO_2 (including $^{238}PuO_2$) is reduced to metal by Direct Oxide Reduction (DOR)
  - Primary reaction takes place in molten $CaCl_2$ at ~900 ^{o}C:

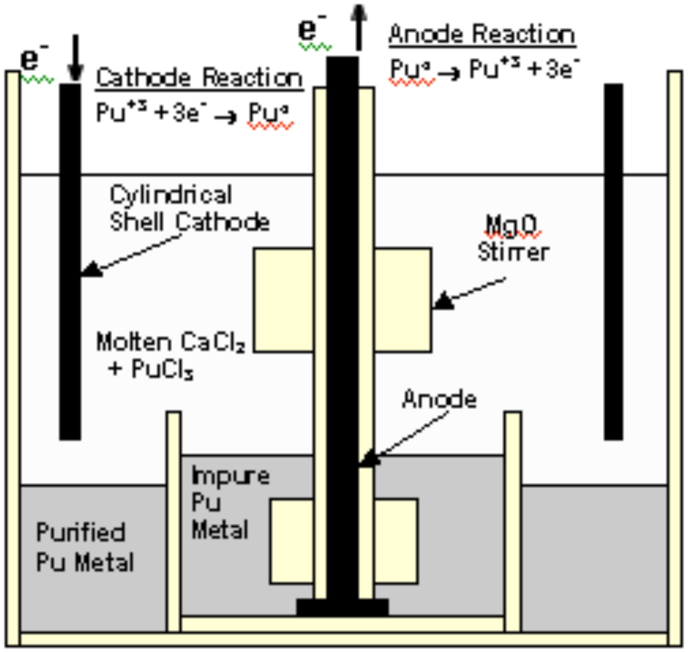
    $$PuO_2 + 2Ca^o \rightarrow Pu^o + 2CaO$$

  - Reaction is spontaneous with ΔG_r^o = -47 kcal/mole PuO_2
- Salt Scrub (SC) is a procedure similar to DOR that is used after DOR to recover any Pu or PuO_2 left behind in the $CaCl_2$ salt phase

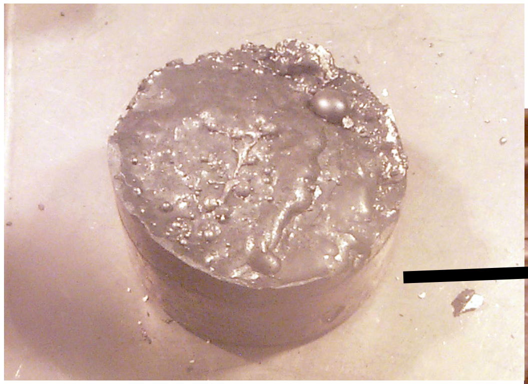
DOR Product Metal Button

Daughter decay products and other impurities are removed from the metal by electrorefining

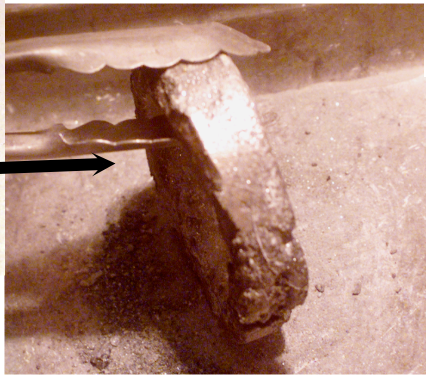
- Electrorefining is used to separate plutonium from impurities by transferring plutonium ions through a molten electrolytic bath using a voltage that is selective for plutonium.
- The purified plutonium metal is produced in a ring shape.



The electrorefining cell operates at 900°C and uses a $CaCl_2$ electrolyte



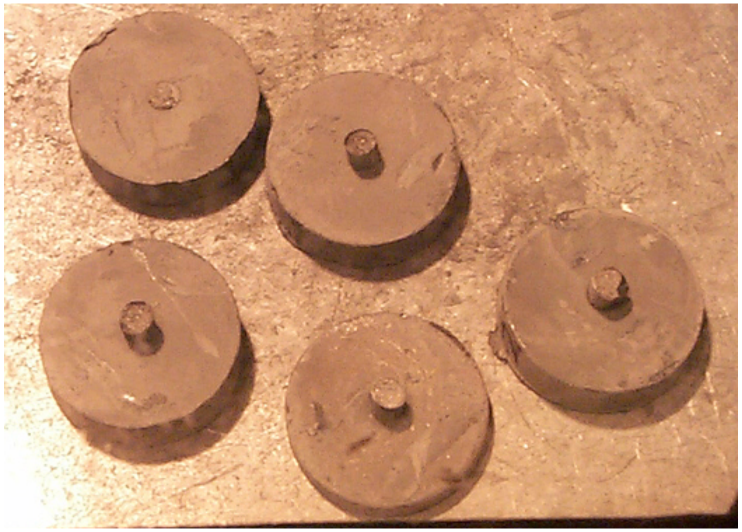
Cast Anode for Electrorefining Feed



Electrorefined product

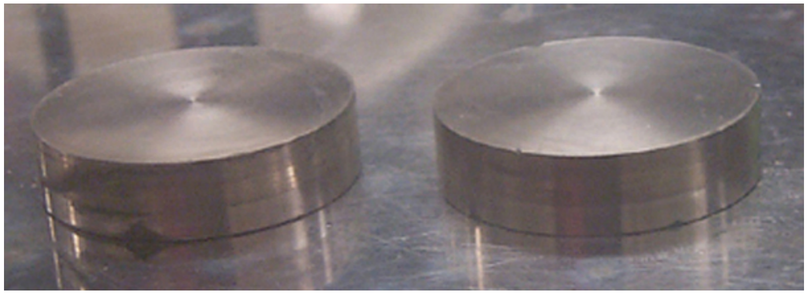
“Cookies” are cast in a graphite mold

- Mold
  - LANL/LLNL design
  - Yttria coated graphite
  - One to ten cookies can be cast in each casting operation
- Cookie dimensions: 1.5” diameter by 3/8” height
- Cookie weight: ~179 grams
- Cast cookie surface is machined to remove excessive graphite and yttria



The spiked alloy is cast into desired shapes

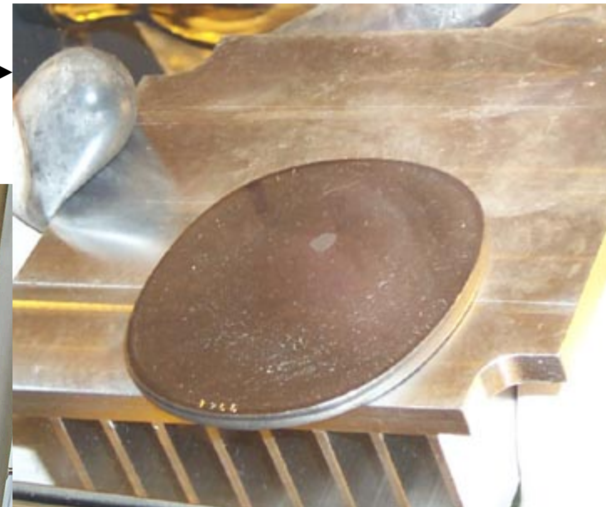
- The Pu alloy is cast into a Y_2O_3-coated graphite (for “cookies”) molds.
- The “cookies” mold is held at 700 ^oC. After casting, the mold temperature is lowered and held for an hour to stabilize the δ-Pu phase, and then cooled.
- The graphite “cookie” molds are a stack of up to 10 "cups." The cups have a hole in the bottom for flow through of the pour. Each cup forms a Pu alloy cookie, 1.5” in diameter by 3/8” high.
- After casting, the molds are broken and the cookies removed.
- The cookies are now reannealed for an hour. They are then rolled to thin the cookies into 3.2 mm thick discs. After rolling, the discs are annealed for 12 hours.



Surface Machined “Cookies”

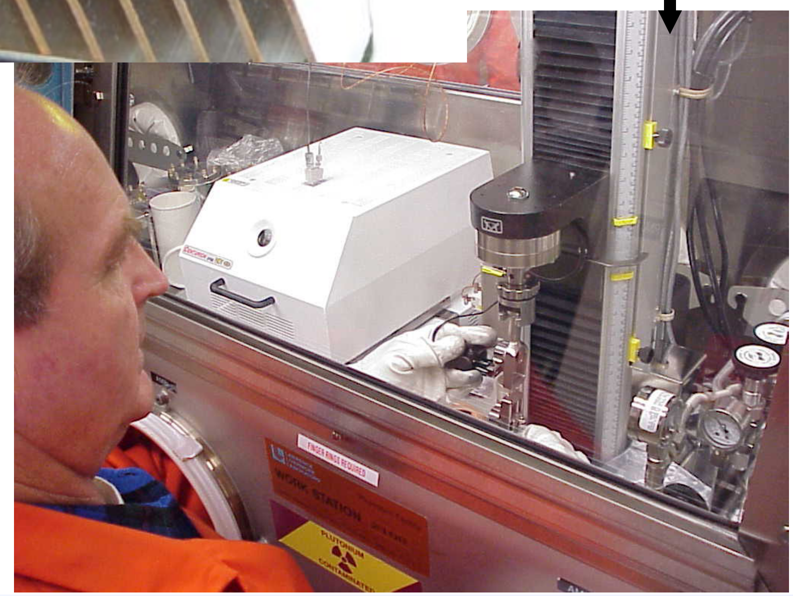
Cast cookies are rolled to disks and annealed in a furnace

Rolled Disk

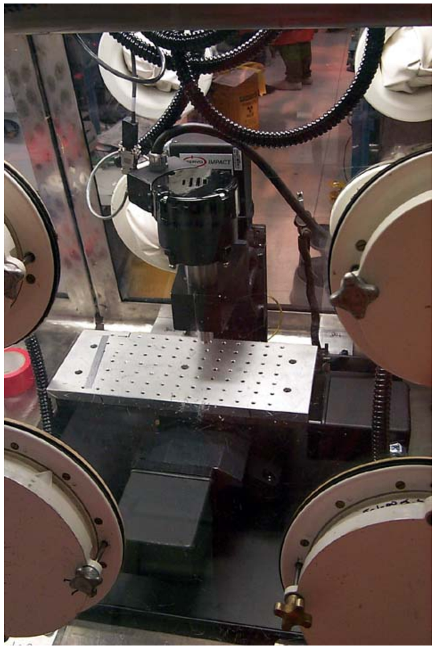


Disk Roller

Annealing Furnace

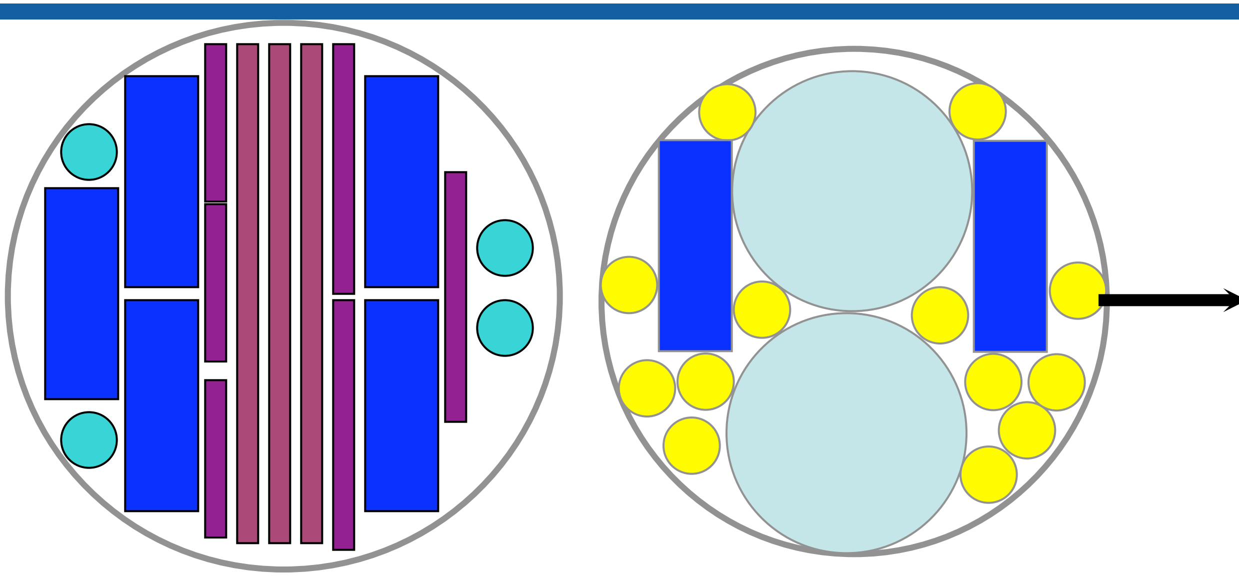


Test specimens are fabricated in a glove box dedicated to ^{238}Pu operations



- Servo “Impact” CNC mill
- CNC Omni-Turn Lathe
- Struers Minitom cutoff saw
- Heidenheim height gage
- Incubator
- Granite block for lapping

Several different cutting patterns are used to obtain desired test specimens



Specimens are rough cut from rolled disk with CNC Mill

= Dilatometer Sample

= compression

= TEM

= Dilatometer Sample

= Dilatometer Sample

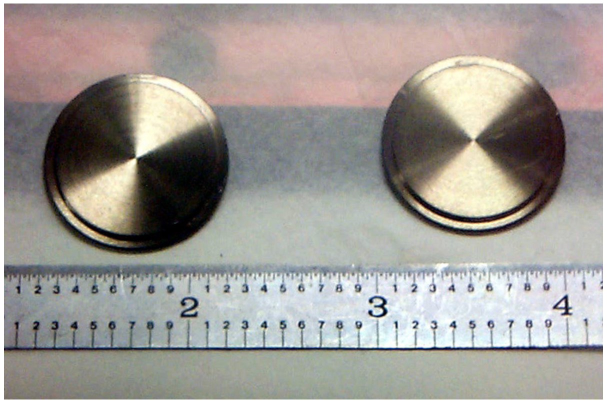
= Tensile

= Dynamic Testing

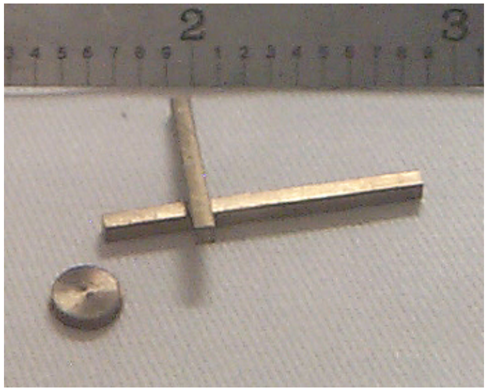
A CNC lathe is used for final machining of test specimens

Density

Tensile Test Specimen



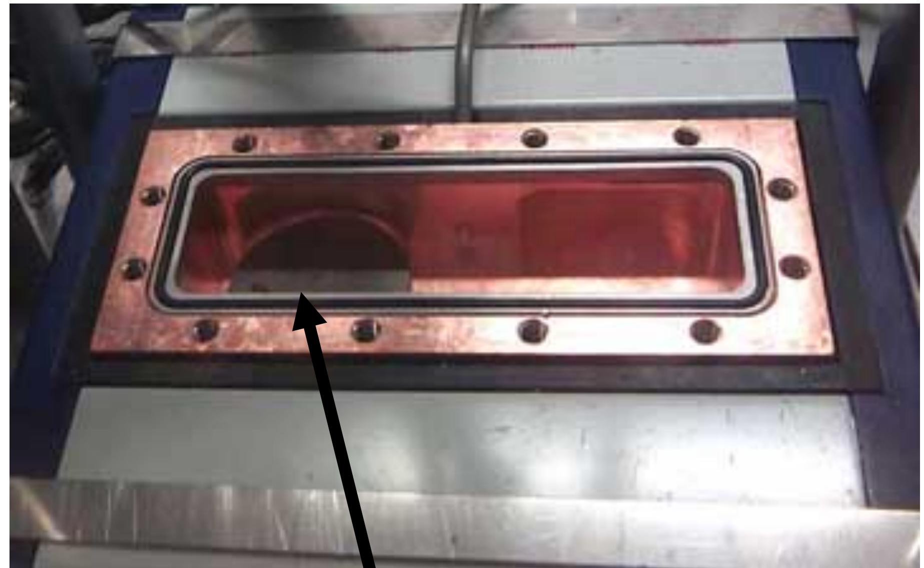
Dynamic Testing



Dilatometer

Test Specimens

Annealed disks and test specimens are stored in an incubator at 50°C



- Techne Dri-block
- Evacuated and backfilled with high purity helium

Rolled and annealed

7.5 wt% ^{238}Pu disk

50°C was chosen as the annealing temperature because it was determined that this was optimum for obtaining a balance between defect generation and annihilation

Analytical data on prepared alloys are within desired range

Element	Reference Alloy #1	Spiked Alloy #1	Spiked Alloy #2	Units
Al	ND	<20	ND	ppm
V	D	D	D	ppm
Cr	D	D	D	ppm
Mn	ND	ND	D	ppm
Fe	410(30)	404(33)	217(18)	ppm
Ni	D	D	D	ppm
Cu	ND	ND	ND	ppm
Ga	D	D	D	ppm
Y	ND	33(24)	10(11)	ppm
Ta	ND	52(72)	26(3)	ppm
^{232}Th	ND	ND	D	ppm
^{233}U	ND	ND	ND	ppm
^{234}U	3(1)	181(44)	129(10)	ppm
^{235}U	80(2)	9(2)	6(.5)	ppm
^{237}Np	11(1)	66(1)	31(2)	ppm
^{238}UPu	0.013(.005)	7.43(.06)	7.26(.08)	atom%
^{239}Pu	93.8(.1)	86.8(.1)	86.99 (0.05)	atom%
^{240}Pu	6.00(.05)	5.56(.06)	5.54(.05)	atom%
^{241}PuAm	0.174(.005)	0.150(.006)	.142(.002)	atom%
^{242}Pu	0.047(.005)	0.057(.004)	.054(.001)	atom%

We've met the challenge!

- We have made ^{238}Pu-spiked plutonium samples to accelerate the aging behavior
- We have characterized these ^{238}Pu-spiked alloys and they look like reference stockpile material
- Co-Authors and Collaborators:
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